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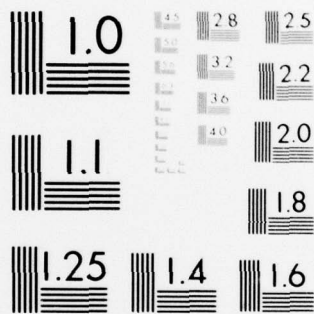
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by

10 S.E./Harris, J./Lukasik, J. F./Young, and L. J. Zych

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# ANTI-STOKES EMISSION AS A VUV AND SOFT X-RAY SOURCE<sup>\*</sup>

S. E. Harris, J. Lukasik,<sup>†</sup> J. F. Young, and L. J. Zych  
Edward L. Ginzton Laboratory  
Stanford University  
Stanford, California 94305

A VUV and soft x-ray light source based on spontaneous anti-Stokes scattering from atomic population stored in a metastable level is described. Unique properties of this source include: narrow linewidth, tunability, linear polarization, picosecond time scale, and quite high spectral brightness. We show how the maximum source brightness, within its narrow linewidth, is that of a blackbody at the temperature  $T$  of a metastable storage level. Experimental results showing laser induced emission at  $569 \text{ \AA}$  and  $637 \text{ \AA}$  from a He glow discharge are described. The use of the anti-Stokes process for direct, internal energy transfer from a storage species to a target species is discussed.

In this paper we discuss some of the properties of a new type of vacuum ultraviolet and soft x-ray light source [1,2]. The source is based on spontaneous anti-Stokes scattering from atomic population which is electrically stored in an appropriate metastable level. The source has several unique properties which include: narrow linewidth, tunability, picosecond time scale operation, linear polarization, and relatively high peak spectral brightness. We will see shortly that this peak spectral brightness corresponds to that of a blackbody at the temperature  $T$  of the storage level. A schematic of the anti-Stokes light source is shown in Fig.1.

Though anti-Stokes scattering is usually described in terms of a spontaneous scattering cross section, it is better for our purpose to describe it in terms of a spontaneous emission rate  $A(\omega)$  induced by the laser pump field  $E_p$  at frequency  $\omega_p$  [3]. This spontaneous emission rate at the vacuum ultraviolet frequency  $\omega$  may be written

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<sup>†</sup> On leave from the Laboratoire d'Optique Quantique du CNRS, Ecole Polytechnique, 91120 Palaiseau, France; recipient of the French-American CNRS-NSF Exchange Award.

$$\begin{aligned}
 A(\omega) &= \frac{\omega^3 |\mu_{13}|^2}{3\pi\epsilon_0 c^3} \left[ \sin^2 \left( \frac{1}{2} \tan^{-1} \frac{\mu_{23} E_p}{\hbar \Delta\omega} \right) \right] g(\omega - \omega_{VUV}) \\
 &= \left( \frac{\omega}{\omega_{31}} \right)^3 A_{31} \left[ \sin^2 \left( \frac{1}{2} \tan^{-1} \frac{\mu_{23} E_p}{\hbar \Delta\omega} \right) \right] g(\omega - \omega_{VUV}) \quad (1)
 \end{aligned}$$

The quantity  $A_{31}$  is the Einstein A coefficient for spontaneous emission from level  $|3\rangle$  to level  $|1\rangle$ . The lineshape  $g(\omega - \omega_{VUV})$  is the convolution of the Doppler- or pressure-broadened linewidth of the  $|1\rangle - |2\rangle$  transition, and  $\Delta\omega$  is  $\omega_{31} - \omega_{VUV}$ .

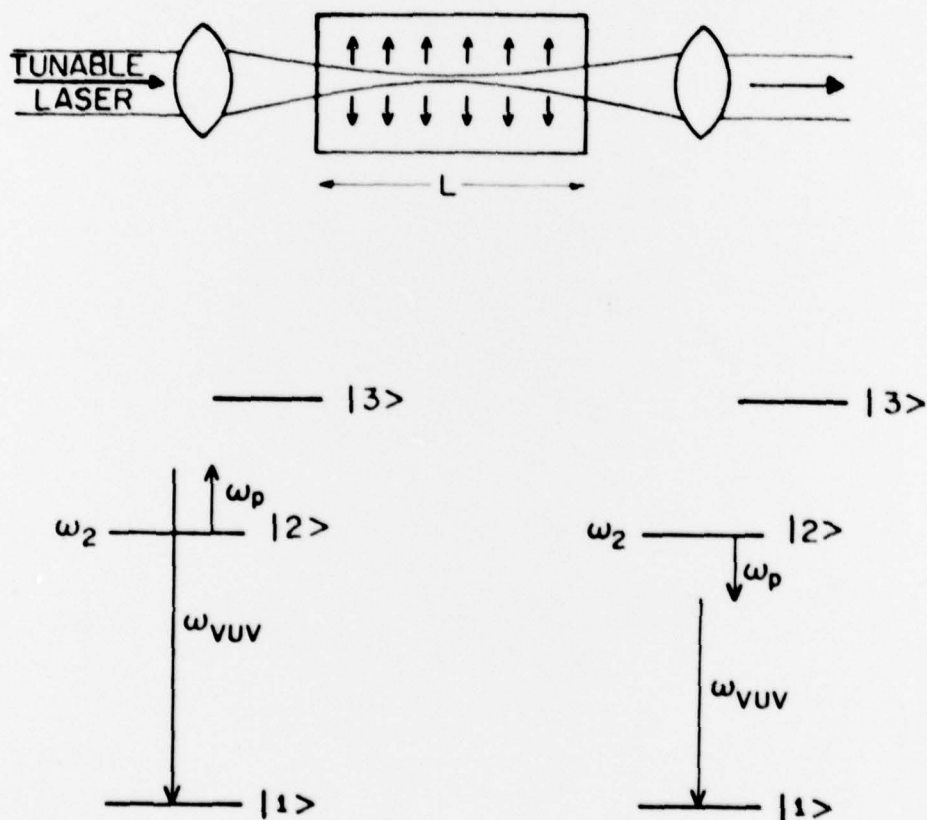


Fig.1 Schematic and energy level diagram for spontaneous anti-Stokes light source. An upper and lower sideband is obtained.

We see from (1) that as the laser pump field becomes large, the anti-Stokes emission rate approaches the Einstein coefficient  $A_{31}$ ; which at fixed oscillator strength  $f_{31}$ , increases as the square of the VUV frequency. The cross section for spontaneous scattering is related to the emission rate by  $\sigma_{sp}(\omega) = \hbar\omega A(\omega)/(P/A)$ , where  $P/A$  is the incident laser power density.



The key to understanding and optimizing this light source is the two-photon absorption which is created at the ultraviolet frequency  $\omega_{VUV}$  in the presence of the laser pump frequency  $\omega_p$ . For the range of laser power densities of interest here, both  $A(\omega)$  and the two-photon absorption cross section  $\sigma(\omega)$  increase linearly with laser power density, and are related to each other in the same manner as are the emission and absorption coefficients for single-photon processes, i.e.,  $\sigma(\omega) = (\pi^2 c^2 / \omega^2) A(\omega)$ .

The brightness of the light source,  $B(\omega)$  photons/(sec cm<sup>2</sup> steradian cm<sup>-1</sup>), is determined by the interplay of the emissive and absorptive processes, and for an infinitely long cylinder of outer radius  $r_0$  is given by [1,2]:

$$B(\omega) = \frac{\hbar \omega^3}{4\pi^3 c^2} \left[ \frac{1}{\exp(\hbar \omega_{21}/kT) - 1} \right] \left\{ 1 - \exp[-\sigma(\omega)(N_1 - N_2)r_0] \right\} \quad (2a)$$

$$\sigma(\omega) = \frac{\pi \omega}{6c^2 \epsilon_0^2 \hbar^3} \left[ \sum_i \left( \frac{\mu_{2i}\mu_{i1}}{\omega_i - \omega_{VUV}} + \frac{\mu_{2i}\mu_{i1}}{\omega_i + \omega_{VUV}} \right) \right]^2 \frac{P_p}{A} g(\omega - \omega_{VUV}) \quad (2b)$$

(mks units).  $T$  is the temperature of the metastable level, i.e.,  $N_2/N_1 = \exp(-\hbar \omega_{21}/kT)$ ;  $\mu_{ij}$  are matrix elements;  $\omega_i$  are the frequencies of the intermediate states; and  $P_p/A$  is the power density of the pump laser.

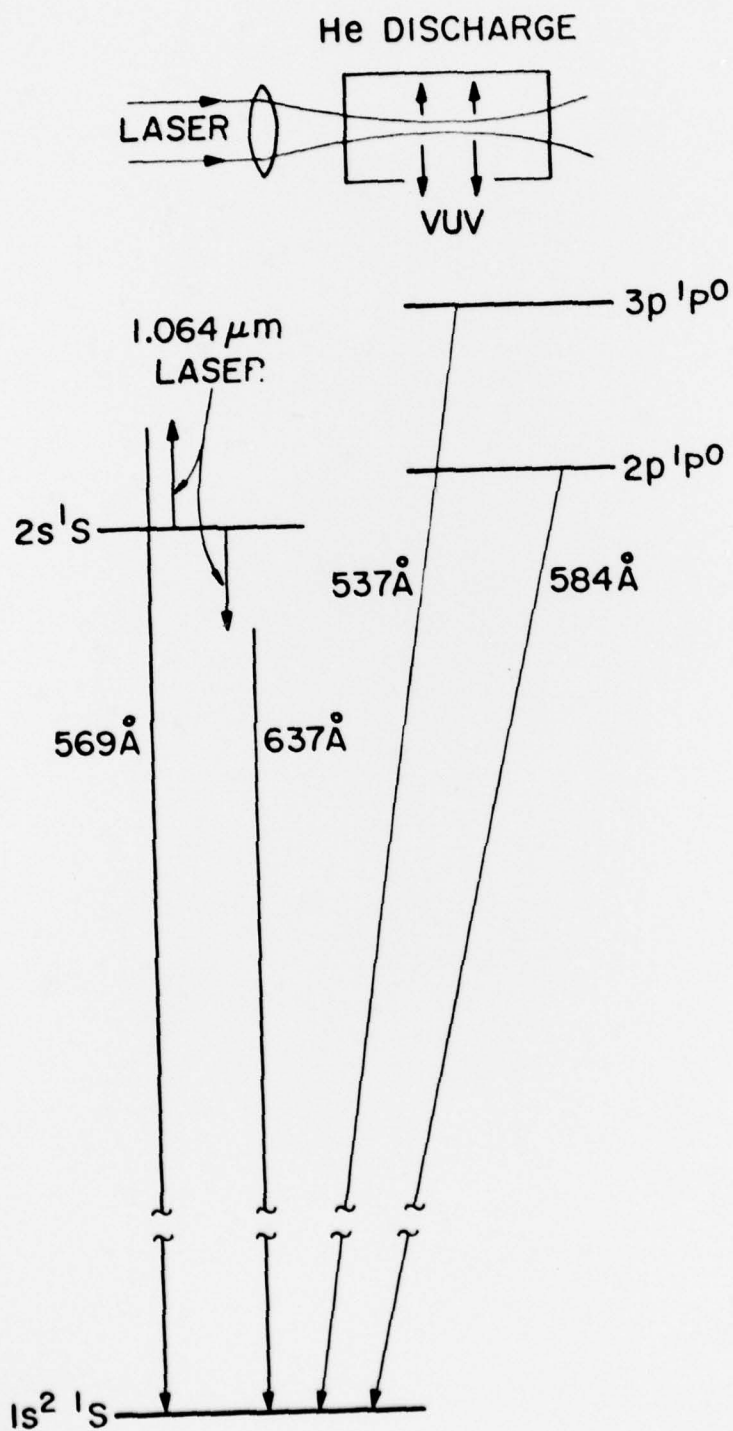
In the (two-photon) optically thin case, i.e.,  $\sigma(\omega)(N_1 - N_2)r_0 \ll 1$ ,  $B(\omega)$  increases linearly with the laser power density and is the same as obtained from the usual spontaneous scattering cross section point of view. As the laser power density is increased and the medium becomes nominally two-photon opaque, i.e.,  $\sigma(\omega)(N_1 - N_2)r_0 = 1$ , the brightness approaches a constant value equal to that of a blackbody radiator at the temperature  $T$  of the metastable level. Once the two-photon opaque or blackbody regime is attained on line center, the primary effect of a further increase in laser power density, cylinder radius  $r_0$ , or ground state density  $N_1$  is to increase the emission linewidth. The total number of emitted photons continues to increase slowly, and the brightness remains constant.

Before proceeding further, we note that anti-Stokes scattering in the VUV has been observed by BRÄUNLICH and LAMBROPOULOS [4], and has been discussed by ZERNIK [5] and VINDOGRADOV and YUKOV [6].

### Experimental Results

In our first experiments [2] on this type of light source a glow discharge was used to store population in the  $2s^1S$  level of He at  $601 \text{ Å} \approx 166,272 \text{ cm}^{-1}$  (Fig. 2). The cw He glow discharge was produced in a 40 cm long quartz tube with a cylindrical hollow cathode and pin anode at opposite ends. Typically, the discharge current was 120 mA and the pressure was about 1 torr. A 0.9 cm long slit was cut through the side wall of the 4 mm ID capillary and served as an input slit for the VUV spectrometer. An actively mode-locked Nd:YAG oscillator-amplifier system produced a train of mode-locked pulses, each with a pulse length of  $\sim 100$  psec. Approximately 10 pulses occurred within the half-power points of the train envelope. The laser was propagated down the discharge capillary tube and focused to an area of about  $3 \times 10^{-4} \text{ cm}^2$  and a

confocal parameter  $b \cong 5$  cm parallel to the input slit.



**Fig.2** Energy level diagram for laser induced emission in He.

The detection system consisted of a spiraltron, a fast preamplifier, a pulse height discriminator, a coincidence gate, and a counter. The coincidence gate was set to a width of 90 nsec overlapping the laser pulse train. Typically,  $10^2$  counts were registered per minute with a signal-to-noise ratio of about 100. The relatively low count rate was a result of the  $3 \times 10^{-9}$  laser duty cycle and the  $2.5 \times 10^{-8}$  ratio of detected photons to total photons generated. Since the source linewidth, even far into the blackbody regime, was well below the resolution of the spectrometer, the observed count rate was proportional to the integrated brightness,  $\tilde{B} \equiv \int B(\omega) d\omega$ .

Using this system we observed laser induced emission at 569 Å, 637 Å, and 591 Å, as well as He resonance line emission at 584 Å, 537 Å, 522 Å, and 516 Å. The emission at 591 Å resulted from anti-Stokes scattering from the He  $2s^3s$  level, and had an intensity of about 1/50 of the 569 Å radiation. Figure 3 shows the relative integrated brightness of the 569 Å radiation as a function of laser peak power density for three He pressures. The points represent the average of 5 one-minute counting intervals. The solid curves are theoretical calculations of  $\tilde{B}$ . As described in [2], the curves were drawn using a value of the two-photon absorption coefficient of  $\sigma(\omega_{VUV}) = 1.8 \times 10^{-26} (P_p/\lambda) \text{ W/cm}^2$ . The magnitude of each theoretical curve was determined by a least squares fit to the experimental points. Our results indicate that the ratio of metastable population to ground state population,  $N_2/N_1 = 2.6 \times 10^{-5}$ , which corresponds to a temperature  $T = 22,700^\circ\text{K}$ . These numbers were independent of pressure in this range to within  $\pm 5\%$ . At the highest power density and pressure of Fig. 3, the two-photon source had a laser induced optical depth of  $r_0 N_1 \sigma(\omega_{VUV}) \sim 7$ , well into the blackbody regime.

The relative intensities of the laser induced emission and the He resonance lines are compared in Table I for a pressure of 1.6 torr and a laser power density of 600 GW/cm<sup>2</sup>. The instantaneous count rate was calculated from the accumulated count using the laser repetition rate and either the coincidence gate aperture time (for the resonance lines) or the effective 1 nsec total laser on-time. In order to estimate the brightness we calculated the linewidth of the 537 Å and 584 Å resonance lines for our geometry and pressure. Based on a Voigt profile, these are 3.2 cm<sup>-1</sup> and 5.6 cm<sup>-1</sup> respectively. The linewidth of the laser induced emission for these operating conditions was calculated as 1.3 cm<sup>-1</sup> at 569 Å and 1 cm<sup>-1</sup> at 637 Å. The second row of Table I also includes a geometrical factor of 2 to account for the larger effective radiating area of the resonance line source. Thus, we estimate that the peak induced emission at 569 Å is 140 times brighter than the strongest He resonance line. As a result of its greater detuning from the intermediate  $2p^1P^0$  level, the brightness of the 637 Å emission is about 7 times smaller than that of the 569 Å emission.

One of the key properties of a laser induced two-photon radiator is that its geometry is dominantly controlled by the pumping laser beam, instead of by the geometry of the discharge. This allows a two-photon radiator to have a temperature characteristic of the interior of a plasma or discharge. In a glow discharge similar to ours, in the interior, electron collisions cause the population of the  $2p^1P$  level to be within a factor of three of the  $2s^1S$  population. However, it is the exterior  $2p^1P$  level atoms which to a large extent determine the temperature of the single-photon 584 Å radiator. As a result of the fact that these atoms are continuously radiating, as well as due to the lower electron density and temperature near the walls, their



temperature may be significantly lower; thereby probably accounting for the factor of  $10^4$  in relative brightness which we have observed. The attenuation and self-reversal of single-photon radiators which results from cold atoms, is also avoided in the two-photon radiator.

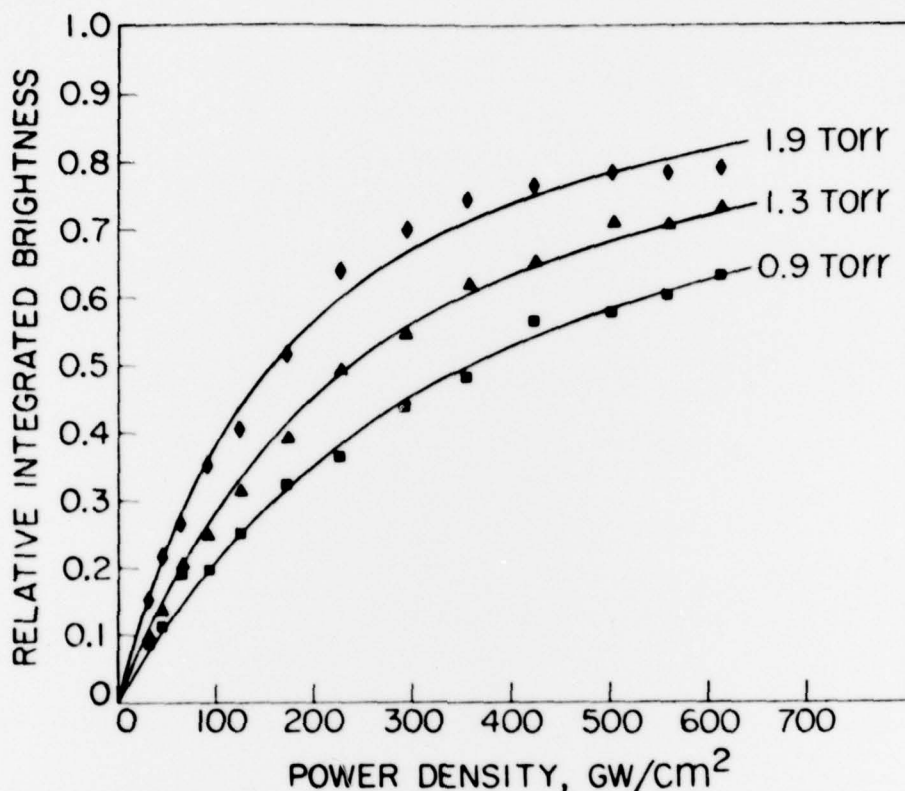


Fig. 3 Relative integrated brightness at  $569 \text{ \AA}$  as a function of  $1.06 \text{ }\mu\text{m}$  laser peak power density. The theoretical curve at each pressure was determined by numerically integrating (2); the magnitude was determined by a least squares fit to the experimental points at that pressure.

#### Flashlamp Applications

One of the uses of this type of light source may be as a flashlamp for short wavelength lasers. To avoid the inefficiencies of short wavelength optics, it may be best to mix the target or lasing species directly with the lamp species. For example, neutral potassium at a density of perhaps  $10^{14}$  atoms/cm<sup>3</sup> might be mixed with He at a density of about  $10^{19}$  atoms/cm<sup>3</sup>. The mixture would then be heated either electrically or by a CO<sub>2</sub> laser beam. At an appropriate time, an incident tunable laser pulse would cause the generation of spontaneous anti-Stokes radiation. This radiation would be absorbed by the neutral potassium, and cause the production of excited-state K<sup>+</sup>. A simplified energy level diagram for this type of interaction is shown in Fig. 4. As shown here, the anti-Stokes source would be tuned to an energy of  $172,732 \text{ cm}^{-1}$  so as to cause an inner shell transition from the  $3p^6 4s$  level to the  $3p^5 4s 4d$  level. A second laser beam of energy greater than  $28,739 \text{ cm}^{-1}$  would carry this excited electron into the continuum causing the formation of the excited  $3p^5 4s$  K<sup>+</sup> ion. By tuning the anti-Stokes source to a discrete

Table 1 Comparison of resonance line radiation and laser induced emission at 1.6 torr and 600 GW/cm<sup>2</sup>.

	Resonance Lines		Laser Induced Emission	
	537 Å	584 Å	569 Å	637 Å
Instantaneous Count Rate 10 <sup>6</sup> Counts/Sec	0.8	32.0	544	57
Estimated Peak Brightness* 10 <sup>15</sup> $\frac{\text{Photons}}{\text{sec cm}^2 \text{ sr cm}^{-1}}$	0.014	0.33	46	6.3

\* The time averaged value is obtained by multiplying by the laser duty cycle of  $3 \times 10^{-9}$ .

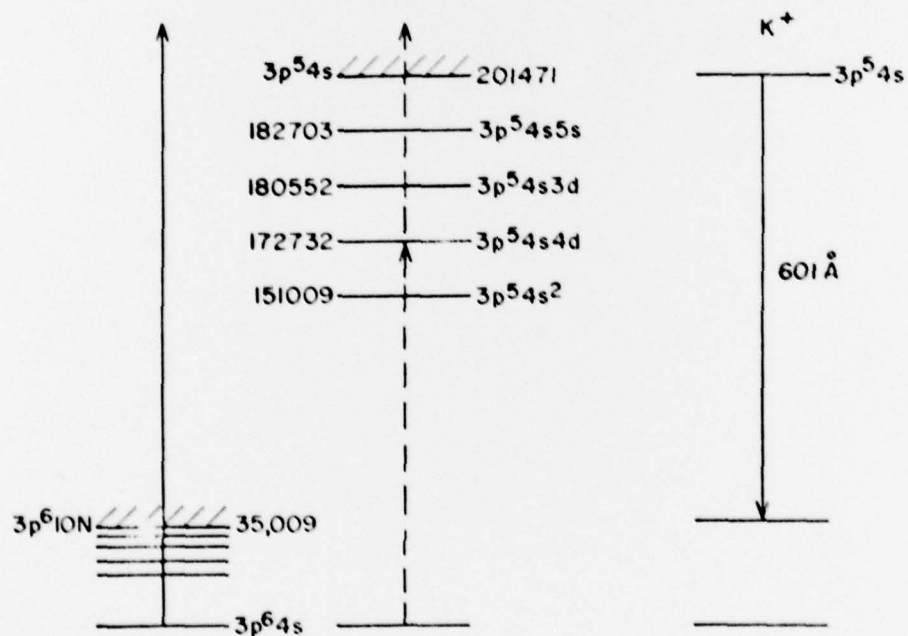


Fig. 4 Simplified energy level diagram for anti-Stokes pumping of K. The (left) solid arrow shows direct pumping to the continuum of an inner shell electron. The dashed arrows show two-photon pumping via an intermediate state.

intermediate state such as shown here, instead of tuning it directly into the continuum as shown by the solid arrow on the left side of Fig. 4, it should be possible to increase the cross section for absorption of the anti-Stokes radiation by about two orders of magnitude. This, in turn, allows operation at a K density much lower than would otherwise be possible, and mitigates, at least somewhat, the problem of the formation of ground state  $K^+$  formed by collision with free electrons.

We should note that the energy required to cause lasing on the 601 Å line of  $K^+$  is quite small. The calculated gain coefficient is  $(2.1 \times 10^{-13}) N \text{ cm}^{-1}$ , where  $N$  is the density of the potassium ions. A gain of  $e^{10}$  in a path length of 1 m requires an ion density of  $5 \times 10^{11} \text{ atoms/cm}^3$ . Assuming a confocal volume of about  $1 \text{ cm}^3$ , this requires an energy of about 1 μJ. This energy must be deposited in a time short compared to the 0.6 nsec spontaneous decay time of the  $K^+$  ion.

Before going further we should note that there are two problems associated with the K-He combination of Fig. 4. The first of these is the formation of ground state K ions by autoionization from the  $3p^5 4s 4d$  level; the second and possibly more severe problem is the formation of ground state K ions by hot electrons in the discharge. During the afterglow, the density of these ions may rapidly reduce by formation of  $\text{HeK}^+$  molecules.

There is an important advantage to the internal or mixed configuration which, in a sense, allows the blackbody limitation to be overcome: to the extent that the product of the single-photon absorption cross section and density of the target species is greater than the two-photon absorption cross section and (ground state) density of the storage species, the anti-Stokes photons will be absorbed by the target instead of reabsorbed by the generating species. For appropriate conditions, the effective anti-Stokes emission rate may then approach that of (1).

We should also briefly address the question of efficiency. At a detuning of  $100 \text{ cm}^{-1}$  from the 2p resonance line of He the cross section for spontaneous anti-Stokes scattering is  $6 \times 10^{-20} \text{ cm}^2$ , and varies as the inverse square of the detuning from the 2p level. Assuming an excited state He density of  $10^{14} \text{ atoms/cm}^3$ , a 1 m path length, and allowing for the energy conversion gain of a factor of  $3^4$ , the ratio of anti-Stokes power generated to laser power incident is about 2%. This assumes that the media is kept (two-photon) optically thin, or that equivalently, as discussed above, all of the energy is absorbed by the target species; and that the excited state population is not depleted.

In conclusion, the anti-Stokes light source has potential for producing radiation in the VUV and soft x-ray spectral regions with many laser-like properties. These include narrow linewidth, tunability, picosecond operation, and controllable polarization. The source may provide a valuable tool for studying the spectroscopy, fluorescent yield, and autoionizing rates of inner shell transitions. Its use as a pump for VUV and soft x-ray lasers is promising.

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References

1. S. E. Harris, Appl. Phys. Lett. 31, 498 (October 1977).
2. L. J. Zych, J. Lukasik, J. F. Young, and S. E. Harris, "Laser Induced Two-Photon Blackbody Radiation in the VUV," Phys. Rev. Lett. (to be published).
3. E. Courtens and A. Szoke, Phys. Rev. A 15, 1588 (1977).
4. P. Braunlich and P. Lambropoulos, Phys. Rev. Lett. 25, 135 (1970); Phys. Rev. Lett. 25, 986 (1970); and P. Braunlich, R. Hall, and P. Lambropoulos, Phys. Rev. A 5, 1013 (1972).
5. Wolfgang Zernik, Phys. Rev. 132, 320 (October 1963); Phys. Rev. A 133, 119 (January 1964).
6. A. V. Vinogradov and E. A. Yukov, Sov. J. Quant. Elect. 3, 163 (September-October 1973).